Protected Silver Coating for National Ignition Facility Flashlamp Reflectors[1]

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ABSTRACT

A durable protected silver coating was designed and fabricated for use on flashlamp reflectors in the National Ignition Facility (NIF) to avoid tarnishing under corrosive conditions and intense visible light. This coating provides a valuable alternative to bare silver where high reflectance and durability are important long-term requirements. This paper describes a protected silver coating having high reflectance from 400 nm to 1000 nm. An alternate coating design extends the high reflectance down to 300 nm while maintaining high reflectance out to 10,000 nm. The specular reflectance is between 95% and 97% in the visible region and 98% or better in the infrared region.

1. INTRODUCTION

The National Ignition Facility at Lawrence Livermore National Laboratory (LLNL) will be a national center to study inertial confinement fusion and the physics of high energy and pressure. The $1.2 billion project consists of 192 laser beams which will direct more than 500 trillion watts towards a tiny target in a pulse only a billionth of a second long. The laser contains neodymium glass amplifiers which are pumped by xenon flashlamps. This is shown in Fig. 1. The flashlamp energy which does not travel directly to the glass laser slabs is reflected by silver reflectors towards the glass laser slabs. These silver reflectors have curved or diamond cross-sections, or are flat. The laser beam is shown incident at the glass laser slabs at Brewster's angle to reduce Fresnel losses. An adjacent amplifier (not shown) is located below the one shown in Fig. 1. This second amplifier shares half of the flashlamp output from the central flashlamp cassette either directly or after being reflected from the silver reflectors of diamond cross-section. The substrates are aluminum or stainless steel.

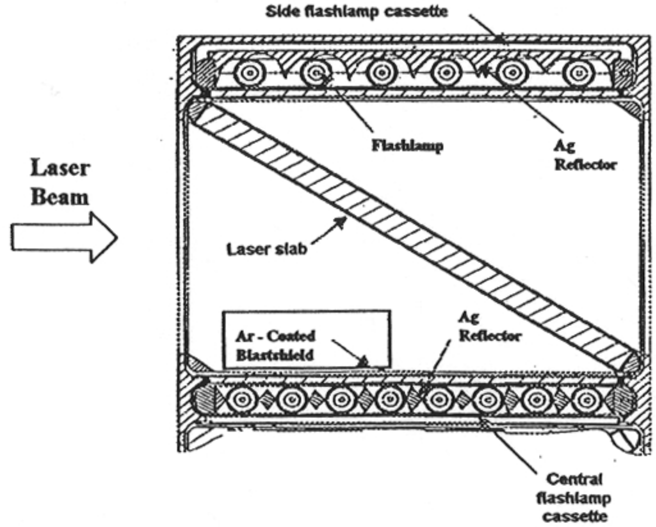

Fig. 1 NIF Amplifier Assembly

1 Presented at the OSA Optical Interference Coatings Topical Meeting, June 7-12, 1998, Tucson, Arizona

Evaporated silver on mirror substrates has several advantages compared to aluminum or stainless steel. It has the highest reflectivity from 400 nm through the infrared and the lowest polarization splitting compared to any other metal. Figure 2 compares the reflectance for silver, aluminum and gold. The disadvantage of bare silver is that it tarnishes under ordinary atmospheric conditions and does not have a high reflectance below 400 nm. There is a minimum reflectance at 320 nm due to a surface plasmon resonance. Aluminum, on the other hand, has a dip in reflectance at 850 nm due to interband transitions, but reflects well below 400 nm.

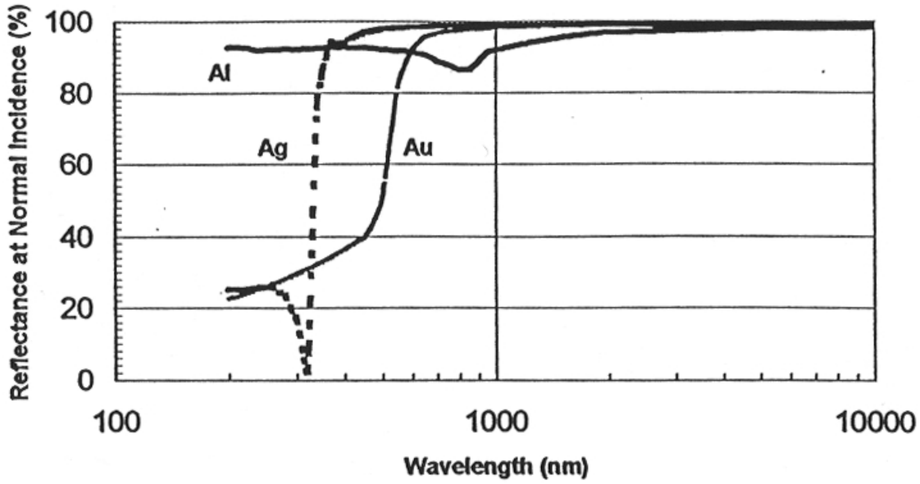

Fig. 2 Reflectance of evaporated metals in the visible and infrared.

Silver is a noble metal which is completely stable in aqueous solutions of any pH as long as oxidizing agents or complexing substances are not present. In the presence of such substances, the high specular reflectivity of silver is degraded by sulfidation, chloridation and oxidation with corrosive chemicals in the atmosphere such as H_2S, O, O^-, H_2O_2, SO_2, CL^-, etc. The corrosion products of silver are Ag_2S, AgCl, Ag_2O, Ag_2SO_4 and Ag_2CO_3 in increasing order of solubility. Since these products form in the thin water layer which is typically present on silver, the most likely precipitate is Ag_2S. Depending on its thickness, an absorbing Ag_2S film can reduce the Ag reflectance to zero. Similarly, the other corrosion films can reduce the reflectance of Ag. AgCl can photolytically decompose in the presence of light leaving metallic Ag which is the basis for some photographic films. The reflectance of an Ag_2S film grown on Ag versus film thickness is shown in Fig. 3.

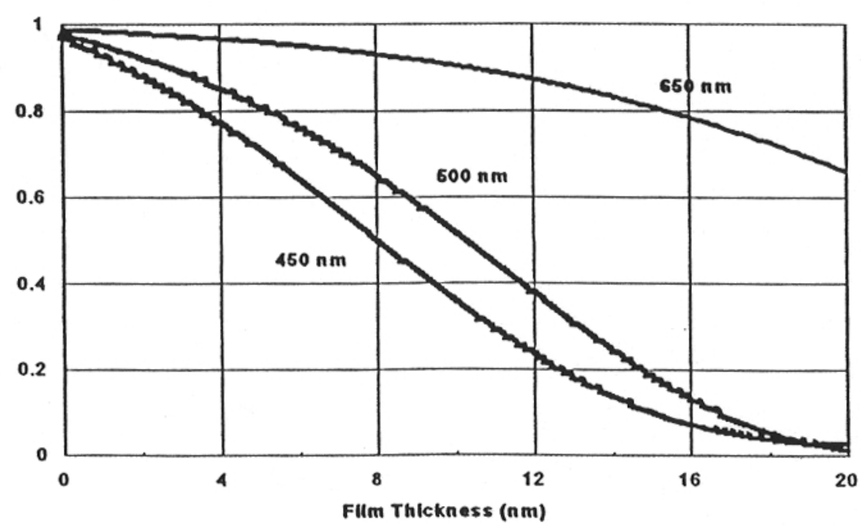

Fig. 3 Reflectance of Ag2S film on Ag versus film thickness

An Ag corrosion experiment showed that the growing film reduces the reflectance more strongly at the shorter wavelengths in complete agreement with the theoretical prediction above (Fig. 3). The film in this case was Ag_2S grown on a sputtered Ag film on an aluminum substrate. Fig. 4 shows the reflectance of this Ag film before and after exposure to 10,000 flashlamp discharges over 2 months in 100% Livermore atmosphere.

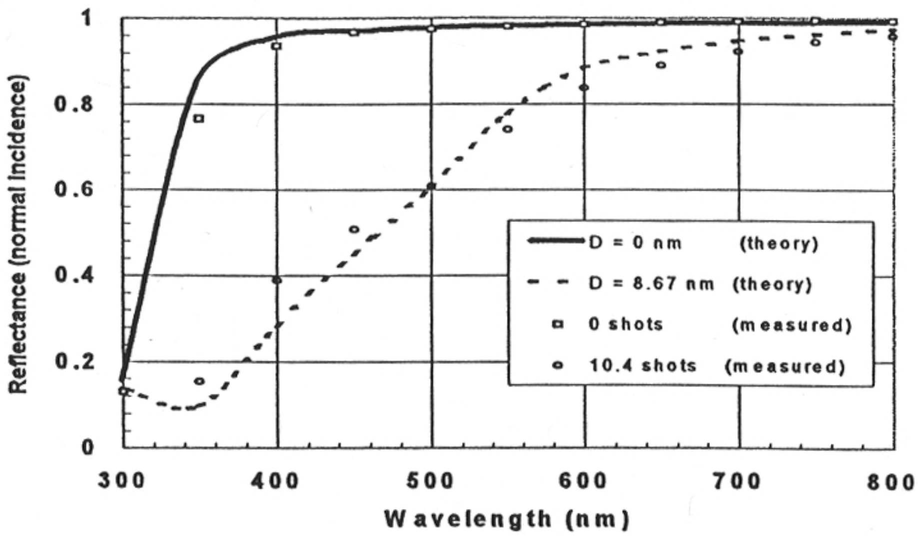

Fig. 4 Reflectance of Sputtered Ag Film Before and After 10^4 Flashlamp Exposures In Normal Livermore Atmosphere for 2 Months: Theory versus Experiment.

The solid curve in Fig. 4 is the theoretical reflectance of Ag calculated from n and k, the squares are the experimental data points for Ag before flashlamp and atmospheric exposure, the circles are the experimental data points after flashlamp and atmospheric exposure, and the broken curve is the best fit for reflectance of an Ag_2S film on Ag. The physical film thickness necessary to achieve this fit is 8.67 nm. This data was used to predict the reflectance of Ag after 10 years which was reduced to 60% based on the Rice[3] model. Of course, this kind of prediction can be very misleading if the film growth model is not correct, and may amount to theoretical target practice. We are conducting an accelerated corrosion test on bare silver to confirm this model or replace it with a better model.

2. THIN FILM DESIGN

The thin film design which may be a candidate for the NIF project is a durable enhanced silver high reflector based on a design presented by J. Wolfe at the OSA meeting in Tucson, Arizona, June 5-9,1995. The design was modified to meet the optical and durability requirements for the NIF project. The coating must meet the following optical specifications:

$$R > 95\% \quad 0° \le \theta \le 60°$$
$$R > 90\% \quad 60° < \theta \le 80°$$

for the wavelength range 400 – 1000 nm. The basic coating design is shown in Fig. 5.

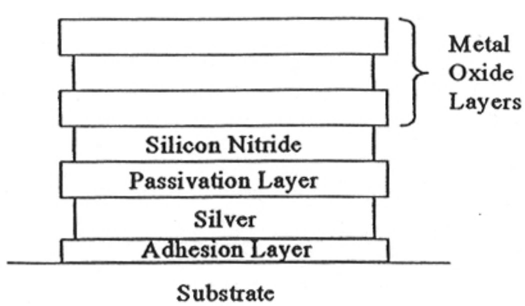

Fig. 5 Coating Design

The adhesion layer provides nucleation sites on the substrate for the sputtered silver , and works in combination with passivation layer to improve mechanical and chemical durability. The passivation layer alloys with the silver and helps in preventing sulfides, chlorides and oxides from reacting with the silver. The dense, hard silicon nitride layer improves mechanical durability and acts as a barrier layer preventing corrodants from reaching the silver. The metal oxide layers are combinations of silica, titania or niobia and serve to increase the reflectance which was slightly reduced by the thin, absorbing silicon nitride layer and passivation layer.

The theoretical optical performance for this design at angles of incidence 20°, 40°, 60° and 80° and 400 – 1000 nm is shown in Fig. 6. This design meets the reflectance specification at non-normal incidence specified for the NIF project.

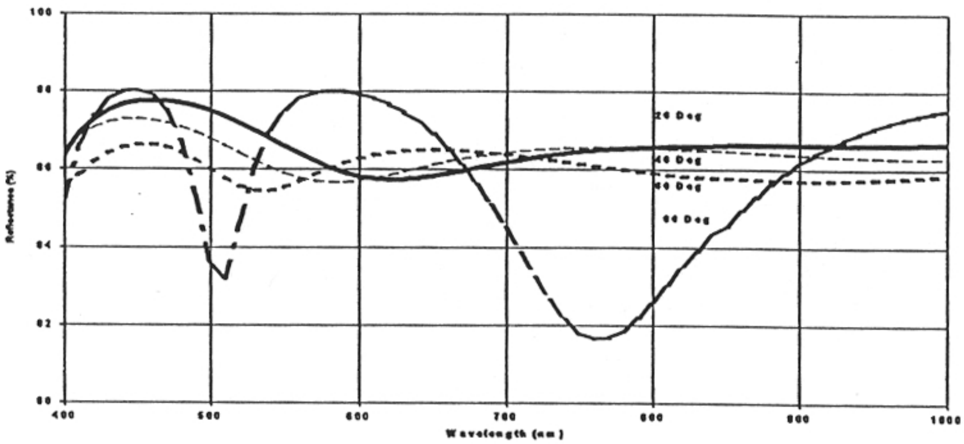

Fig. 6 Theoretical reflectance of coating design at angles of incidence 20°, 40°, 60°, and 80°.

3. PRELIMINARY COATING RUNS

Viratec Thin Films, Inc., in Faribault, Minnesota, provided their R&D facility and expertise for the coating. The design was coated on aluminum and stainless steel substrates using an Airco ILS-1600 sputter coater with a load-lock and DC magnetrons. Single layers of the various coating materials were coated onto separate aluminum substrates and characterized with a Woollam ellipsometer, Alpha Step profilometer, and Perkin Elmer Lambda 9 spectrophotometer. The adjusted design was coated onto diamond-paste polished aluminum substrates, stainless steel substrates, highly polished aluminum foil, and microscope slides overcoated with sputtered aluminum as a base layer. The coated microscope slides were scanned with a Perkin Elmer Lambda 9 spectrophotometer with an absolute reflectance attachment. The resulting scan at near-normal incidence is compared to theory in Fig. 7. The measured specular reflectance agrees to within 1% of the theoretical

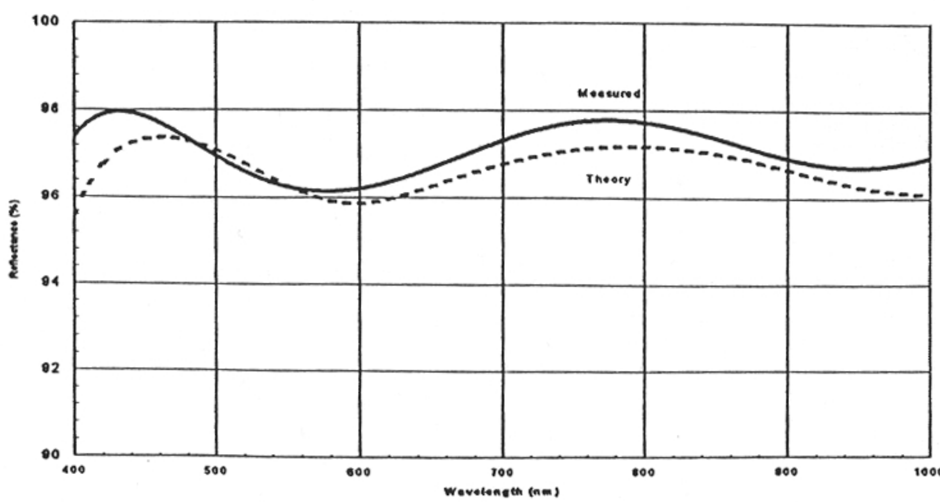

Fig. 7 Comparison of the measured coating reflectance to theory

prediction. Several highly polished aluminum foils 0.030 inch thick and highly polished stainless steel 0.029 inch thick substrates were coated with the design which showed excellent results.

4. ENVIRONMENTAL TEST RESULTS

The coated parts were tested according to Mil Spec 13508C. They successfully passed tape, cheese cloth, and humidity. An additional test was performed in the flashlamp test facility where a coated aluminum sample was subjected to flashes from an intense flashlamp in a normal Livermore atmosphere with naturally-occurring corrodants. After two months testing and 20,717 flashes, there was no detectable change in reflectance of the protected silver. During the same test, bare silver mirrors corroded and either peeled off the substrates or developed a haze on the silver which was probably due to the growth of Ag_2S micro-crystals. Some other over-coated silver mirrors also failed the test.

The reflectance of the successful protected silver mirror before and after exposure to 20,717 flashlamp discharges over 4 months is shown in Fig. 8. The reflectance curves before and after exposure lie on top of one another to within 1%, the accuracy of the measurement. A diamond scribe was used to scratch through the top coating layers which were exposed to normal atmosphere without any apparent corrosion. The silicon nitride layer is thought to be responsible for this mechanical durability.

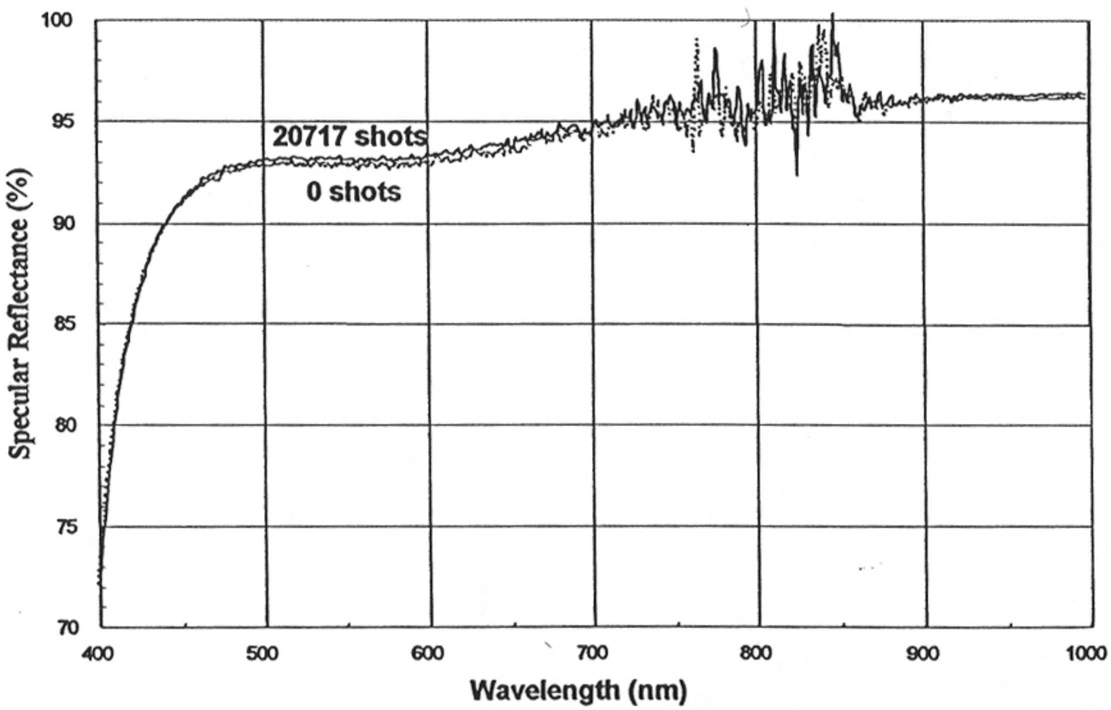

Fig. 8 Reflectance of protected silver mirror before and after 20,717 flashes and 4 months in a corrosive atmosphere.

The reflectance of the same protected silver mirror was measured in the infrared. Fig. 9 shows the reflectance at normal incidence from 3 μm to 13 μm (3000 nm to 13,000 nm). The reflectance is typically very high in the infrared.

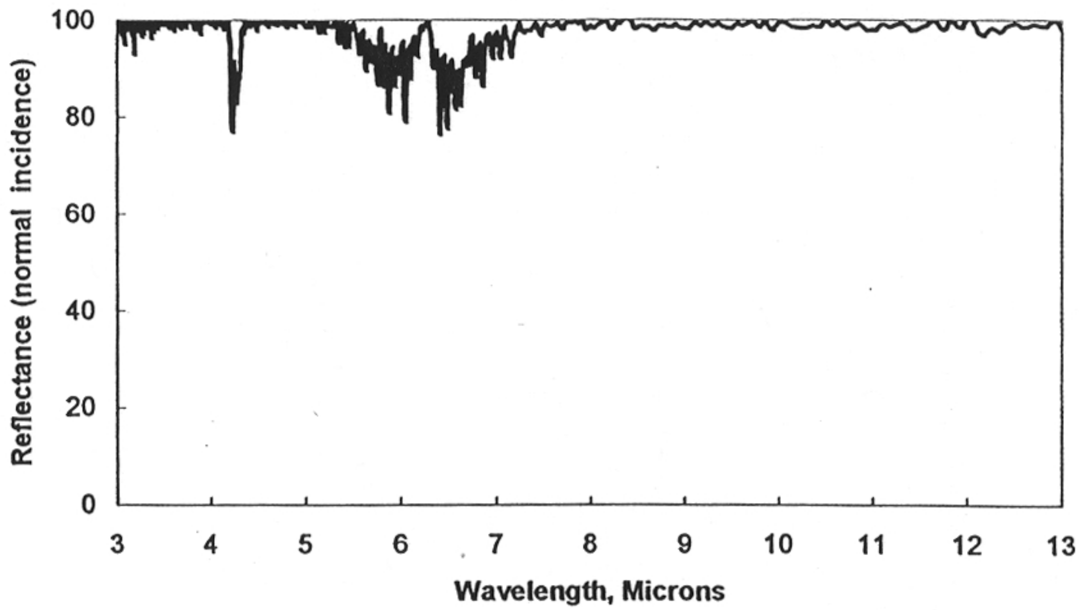

Fig. 9 Measured reflectance of protected silver mirror in the infrared.

5. FUTURE DEVELOPMENT

The future development of protected silver coatings for astronomical mirrors will include extending the reflectance down to 300 nm. The initial coating design shows promise in achieving this goal. We are working towards the goal of preventing tarnishing for at least 30 years under normal atmospheric conditions.

6. CONCLUSIONS

A durable protected silver coating was designed and coated on polished aluminum and stainless steel substrates, aluminum foils and microscope slides. This coating meets the optical performance specifications required for NIF. Initial testing according to mil-spec 13508C indicates that it has the mechanical and chemical durability required for the NIF flashlamp reflectors. Further accelerated testing under flashlamp irradiation while in a corrosive environment will being conducted in order to ascertain the corrosion rates to be expected over the 30-year lifetime of the National Ignition Facility. The results indicate that this durable protected silver coating may be a good candidate for other applications such as astronomical mirrors, high efficiency gratings, copiers, projection TV's, bathroom mirrors, etc. It offers the advantage of durability and high reflectance when compared to aluminum coatings. It will be especially advantageous when extended down to 300 nm for astronomical applications.

7. ACKNOWLEDGMENT

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